Magnetic polarization induced by nonmagnetic impurities in high-T_c cuprates

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The magnetic polarization induced by nonmagnetic impurities such as Zn in high-T_c cuprate compounds is studied by the variational Monte Carlo simulation. The variational wave function is constructed from the eigenstates obtained from Bogoliubov de Gennes mean-field Hamiltonian for the two-dimensional t-J model. A Jastrow factor is introduced to account for the induced magnetic moment and the repulsion between holes and the impurity. A substantial energy gain is obtained by forming an antiferromagnetic polarization covering four or five lattice sites around the impurity. We also found the doping dependence for the induced magnetic moment consistent with experiments.

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Recently a number of experiments, the neutron scattering,[1] nuclear-magnetic resonance (NMR)[2,3] and scanning tunneling microscopy (STM),[4–6] have been carried out to study the impurity effect on the electronic transport and magnetic properties in high-T_c cuprate compounds. These studies provide detailed information about the relationship between magnetism and superconductivity in high-T_c cuprates. The nonmagnetic impurity Zn was found to suppress T_c more strongly than magnetic impurity Ni, even though both replace Cu in the CuO_2 plane.[7] The amazingly accurate measurement of the local density of states (LDOS) by STM (Refs. 4–6) also provides very different spectra for Zn and Ni. The spin dynamics studied by the neutron-scattering experiments reveals that the low-energy spin fluctuations are strongly enhanced near the impurity and the magnetic excitation at the antiferromagnetic wave vector (π,π) disappears with Zn doping in the underdoped region.[8,9] It is interesting to find from the NMR and SQUID experiments that both the nonmagnetic Zn and the magnetic Ni impurities induce a local magnetic moment on Cu sites surrounding the impurity in the normal state. The broadening of ^{63}Cu and ^{17}O NMR lines has been attributed to a distribution of magnetic moments or a spatially inhomogeneous spin polarization extending over several lattice sites around the impurity. On the other hand some experiments[10] found no evidences of the existence of local magnetic moments, at least in the optimum and overdoped samples. More careful theoretical and experimental efforts to examine the magnetic polarization are needed to clarify this issue.

So far most of the theoretical work has been based upon phenomenological BCS-type models with emphasis on understanding of the LDOS. The observed nearly zero-energy-resonance peak near Zn impurity was explained very early by Balatsky and co-workers[11–14] by assuming Zn to be an unitary impurity. Studies[15–20] based upon t-J-type models have also successfully explained the LDOS. There are only few studies[18] about the structure of magnetic polarization induced by the magnetic moment bound to the nonmagnetic impurity and the screening of this moment by other electrons. However, in a recent paper[21] Tsuchiura *et al.* use the Gutzwiller approximation and the Bogoliubov de Gennes (BdG) approach for the t-J model and they find no evidence of the existence of the local moments around the Zn impurity. They also concluded that the electron avoids the impurity instead of being bound to it. A much more careful examination of the effect of a nonmagnetic impurity in the t-J model is needed to resolve the controversy.

Comparing with other phenomenological models, the t-J model has much stronger magnetic correlation and it may lead to a different picture about the magnetic polarization around the impurity. However, previous studies of the t-J model use the BdG approach with or without the Gutzwiller approximation and the non-doubly-occupied constraint imposed by the t-J model is only taken into account on the average or approximately. It very likely underestimates the antiferromagnetic correlation inherent in the t-J model. Another issue that has not been addressed adequately before is the doping dependence of the induced magnetic moment. Very different results reported by NMR experiments[2,7,10] may be related to the doping dependence.

In this paper we will impose the constraint rigorously by using the variational Monte Carlo approach[23] to study the effect of nonmagnetic Zn impurity on the ground state of the t-J model. The ground-state trial wave function is first constructed by assuming d-RVB (resonating-valence-bond) order parameters in the BdG approach. Then the variational wave function is shown to be greatly improved by adding a Jastrow factor to account for the strong magnetic correlation. We found a large energy gain by having an antiferromagnetic polarization around the impurity with a size of about 4–5 lattice sites as observed in ^{63}Cu NMR data[2] in the underdoped region. The significant suppression of the magnitude of the induced moment and its polarization size as doping increases to optimum doping is also consistent with experimental observations.[3,7] In addition, our result also provides a reason to explain the similarity between results[3] measured for Li^+ and Zn^{2+}. Contrary to the work reported in Ref. 21 we show that electrons are always attracted to the impurity. But the effect gets weaker when number of holes increases.

The model we consider is the dilute impurity limit of the two-dimensional t-J model. The interaction between impurities is neglected. $Zn^{2+}(3d^{10})$ has total spin $S=0$ and its second ionization energy is about 18 eV. Near chemical potential the conduction electron is estimated to encounter a

repulsive local potential $U_0 \approx 18.9$ eV (Ref. 24) when it scatters with the Zn impurity. This is much larger than the bandwidth (2 eV) of the $d_{x^2-y^2}$ band of $3d$ Cu^{2+} electrons. Thus, the nonmagnetic impurity Zn can be described roughly by a spin vacancy in the unitary limit. We start from the Hamiltonian,

$$H = -t \sum_{\langle ij\rangle,\sigma} P_G (c^\dagger_{i\sigma} c_{j\sigma} + \text{H.c.}) P_G + J \sum_{\langle ij\rangle} \left(\mathbf{S_i} \cdot \mathbf{S_j} - \frac{1}{4} n_i n_j \right) + \sum_i (U_0 \delta_{i,I} - \mu) n_{i\sigma}, \tag{1}$$

where I labels the site of the impurity. In the standard notation, the $\langle ij \rangle$ means the summation over nearest neighbors and $P_G = \Pi_i (1 - n_{i\uparrow} n_{i\downarrow})$ is the Gutzwiller's projection operator that prohibits double occupancy. Within the mean-field approximation, the BdG equation is derived as

$$\sum_j \begin{pmatrix} h_{ij} & F_{ij} \\ F^\dagger_{ij} & -h_{ij} \end{pmatrix} \begin{pmatrix} u^m_j \\ v^m_j \end{pmatrix} = E_m \begin{pmatrix} u^m_i \\ v^m_i \end{pmatrix}, \tag{2}$$

where

$$h_{ij} = -\left(t\delta + \frac{1}{4} J \chi_{ij} \right) + (U_0 \delta_{I,i} - \mu) \delta_{i,j}, \tag{3}$$

$$F_{ij} = -\frac{1}{2} J \Delta_{ij}. \tag{4}$$

Here u^m_i and v^m_i are the Bogoliubov amplitudes corresponding to the eigenvalue E_m; χ_{ij} and Δ_{ij} are the bond and RVB order parameters defined by $\chi_{ij} = \Sigma_\sigma \langle c^\dagger_{i\sigma} c_{j\sigma} \rangle$ and $\Delta_{ij} = \langle c_{i\downarrow} c_{j\uparrow} - c_{i\uparrow} c_{j\downarrow} \rangle$, respectively; δ is the hole density. They are determined self-consistently by

$$\chi_{ij} = 2 \sum_m u^{m*}_i v^m_j, \tag{5}$$

$$\Delta_{ij} = -2 \sum_m u^{m*}_i v^m_j, \tag{6}$$

$$\delta = \frac{1}{N} \sum_{m,i} (|u^m_i|^2 - |v^m_i|^2). \tag{7}$$

The solution found at zero temperature had already been shown by several groups[19,20] to have a nearly zero-energy resonance for the LDOS when U_0 is very large compared to J or t. The order parameters Δ_{ij} near the impurity are suppressed and a small component of s-wave pairing is induced. In the slave-boson mean-field theory,[22] the magnetic correlation obtained is overestimated. The simplest way to correct this deficiency is to use the eigenvectors obtained by BdG equations to construct a variational wave function with the projection operators rigorously imposed. For the uniform case[23] a similar method has been used successfully.

Following the work by Yokoyama and Shiba[25] and Himeda *et al.*,[26] we write this trial wave function for the ground state in terms of a Slater determinant of $N_e/2$ dimension,

$$|\phi\rangle = P_G \left(\sum_{ij} (U^{-1}V)_{ij} c^\dagger_{i\uparrow} c^\dagger_{j\downarrow} \right)^{N_e/2} |0\rangle, \tag{8}$$

where U and V in Eq. (8) are the matrices of u^m_i and v^m_j, respectively. Without the Gutzwiller's projection operator, this wave function is exactly the same as BdG ground state but with fixed N_e electrons. The relation of this wave function with superconductivity in the absence of impurity was discussed in Refs. 22 and 24. Most properties calculated with or without the Gutzwiller's projection operator are quite similar as shown by Zhang *et al.*[27] However, the spin-spin correlation calculated by BdG [Eq. (2)] is very much smaller than by Eq. (8).

It should be noted that the trial wave function in Eq. (8) is a paramagnetic RVB state without the antiferromagnetic long-range order (AF LRO). In a uniform system without impurity at low doping, $\delta < 0.06$, this state is unstable[28] with respect to the AF LRO. To take into account AF LRO, we could either add a Jastrow factor,[28] such as $\exp[-h_u \Sigma_i (-1)^i S^i_z]$, to modify the trial wave function or we could include spin-density wave order parameter[29,30] to the original BdG equations. Since both approaches obtain almost identical results, we shall use a Jastrow factor here.

In addition to the issue of AF LRO at low doping, we are also concerned with the lack of consideration of strong correlation in the mean-field theory of BdG equations. Use of Gutzwiller's approximation[21] in BdG would improve the situation but it still may not be enough. When the no-double-occupancy constraint is included exactly, we could examine the issue of attraction[21] or repulsion[16–18] of holes by the impurity more accurately. Hence we introduce a Jastrow factor to reflect the influence of the impurity on the nearby hole distribution and magnetic polarization. This new trial wave function is

$$|\psi_I\rangle = \exp\left[-\sum_i \left((-1)^i h_i S^z_i + \frac{\lambda(1-n_i)}{R_i} \right) \right] |\phi\rangle, \tag{9}$$

where $R_i = \sqrt{(x_i - x_I)^2 + (y_i - y_I)^2}$ is the distance from the impurity site denoted by I. The first term in the exponent in Eq. (9) introduces a spatial dependent staggered magnetic field, which consists of two terms, $h_i = h_u + (h_0)/(R_i)$. h_u provides a uniform AF LRO at low doping with or without the impurity. h_0 is used to describe the enhanced AF correlation effect around the impurity. This enhancement will repel holes away from the impurity. Hence we include the second term associated with λ for this repulsion. Notice that if λ is negative, then the hole is attracted to the impurity and the electron is repelled from it. The values of h_u, h_0, and λ are determined by minimizing the variational energy. In Eq. (9) we have chosen $1/R$ form to simulate the extent of the spin polarization around the impurity. We have examined several other functional forms and results are about the same as long as it covers a substantial region around the impurity.

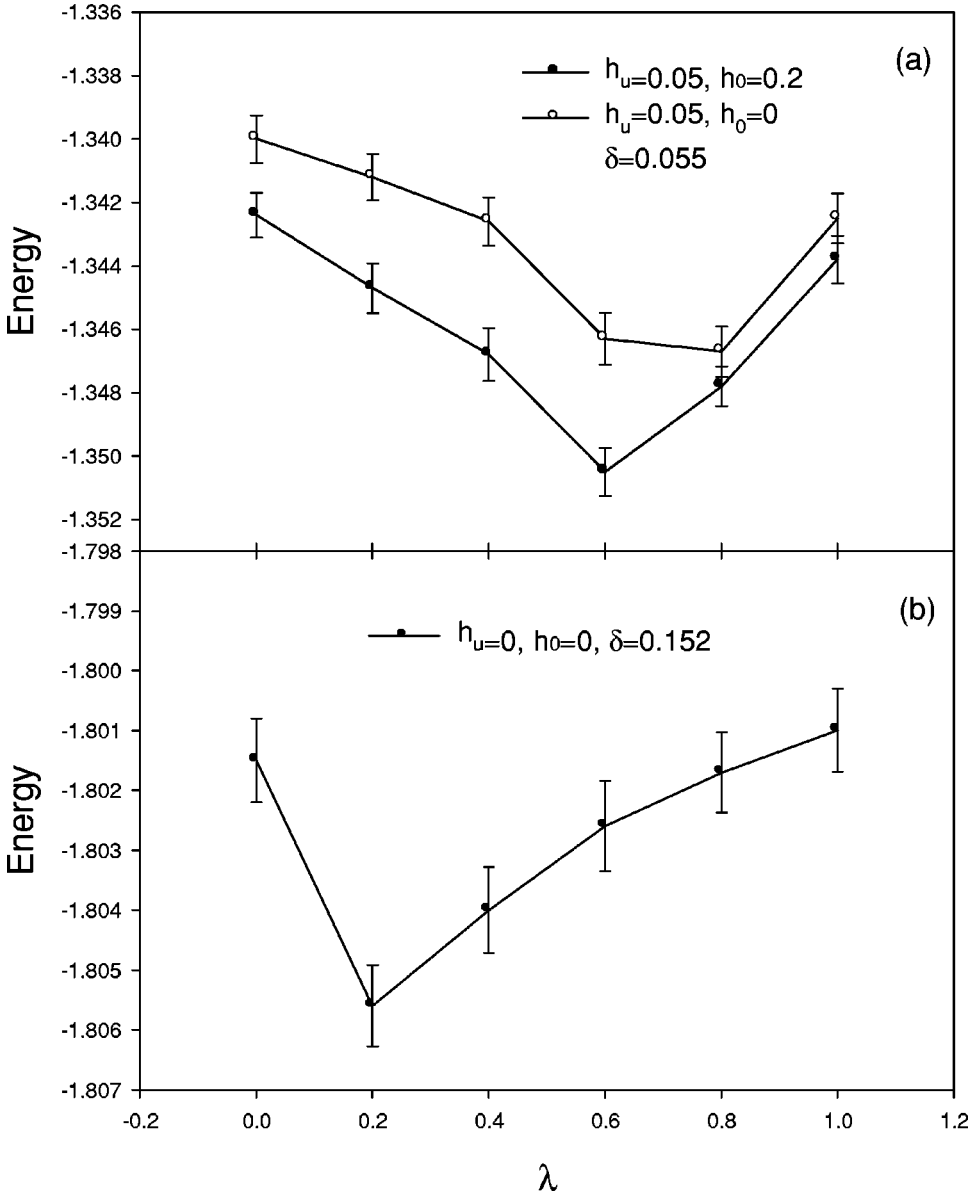

FIG. 1. Variational energies plotted as a function of the impurity-hole repulsion parameter λ. Figures 1(a) and 1(b) are for different values of the parameters as speicifed in the figure.

Below we will report mostly on the results obtained from $|\psi_I\rangle$ with $1/R$ form. Then we will also show that similar results are obtained with a different trial function $|\psi_I'\rangle$ using $1/R^2$ form. The latter has been previously shown by Khaliullin *et al.*[18] to be the spatial distribution of the impurity-induced moment.

Our attention is also focused on the spatial magnetic polarization near the impurity. Without loss of generality the impurity is supposed to be situated at the center of the lattice. Thus, we can use the periodic boundary condition for the numerical calculation. For the 8×8, 12×12, and 16×16 lattice sizes we find that the spin cloud induced by the impurity extends only several lattice sites and all the quantities we are concerned with, including the local magnetization and the spin-spin correlation function, have no qualitative and significant changes with the change of the lattice size. This is because the lattice sizes we used are large enough for the polarized spin cloud. Here we present the numerical results

TABLE I. Optimal ground-state energy per site as a function of hole density for two trial wave functions:$|\phi\rangle$ and $|\psi_I\rangle$. The third row lists their total energy difference.

Doping δ	0.028	0.055	0.083	0.111	0.139	0.152
$\|\phi\rangle$	-1.207 ± 0.002	-1.336 ± 0.004	-1.475 ± 0.002	-1.611 ± 0.003	-1.738 ± 0.003	-1.801 ± 0.002
$\|\psi_I\rangle$	-1.224 ± 0.001	-1.351 ± 0.001	-1.487 ± 0.002	-1.623 ± 0.003	-1.748 ± 0.002	-1.806 ± 0.001
ΔE	-2.4 ± 0.4	-2.3 ± 0.7	-1.7 ± 0.6	-1.7 ± 0.9	-1.4 ± 0.7	-0.7 ± 0.3

TABLE II. Optimized variational parameters for $|\psi_I\rangle$. The values in the parenthesis are for the clean system without impurity.

Doping δ	0.028	0.055	0.083	0.111	0.139	0.152
h_u	0.1(0.1)	0.05(0.05)	0(0)	0(0)	0(0)	0(0)
h_0	0.3(0)	0.2(0)	0.05(0)	0.03(0)	0(0)	0(0)
λ	0.8(0)	0.6(0)	0.4(0)	0.4(0)	0.2(0)	0.2(0)

obtained for a 12×12 lattice in the zero temperature limit with $t/J=3$ and $U_0=100J$. In this paper J is our basic energy unit. We solve self-consistently the BdG equations and obtain the order parameters χ_{ij}, Δ_{ij}, and the BdG amplitudes U and V. The pairing order parameters Δ_{ij} can be decomposed into extended s-wave and d-wave components as $\Delta_d(i)=\frac{1}{4}[\Delta_x(i)+\Delta_{-x}(i)-\Delta_y(i)-\Delta_{-y}(i)]$ and $\Delta_s(i)=\frac{1}{4}[\Delta_x(i)+\Delta_{-x}(i)+\Delta_y(i)+\Delta_{-y}(i)]$. The d-wave component is suppressed around the impurity site and it induces a small s-wave pairing component that is consistent with other group's results.[20,31] In principle, the Jastrow factor introduced could modify the distribution of the order parameters. In practice, tuning the values of the order parameters around the solutions of the self-consistent BdG equations has little effect on the physical quantities discussed below, except a slightly lower ground-state energy is obtained. After obtaining the BdG solution and matrices U and V, we carry out the VMC simulation to determine the optimized ground-state energy. About 10^5 samples were used in each MC simulation to measure the physical quantities. Since there are three variational parameters: h_u, h_0, and λ, the calculation to find the optimal solution is quite involved. Here we only report on the main results. In Fig. 1 we show energy per site as a function of λ for two doping concentrations. In Fig. 1(a) for doping concentration $\delta=0.055$ and $h_u=0.05$, results for $h_0=0.2$ (solid circles) and $h_0=0$ (open cirlces) are compared. Figure 1(b) shows that the lowest energy for $\delta=0.152$ is

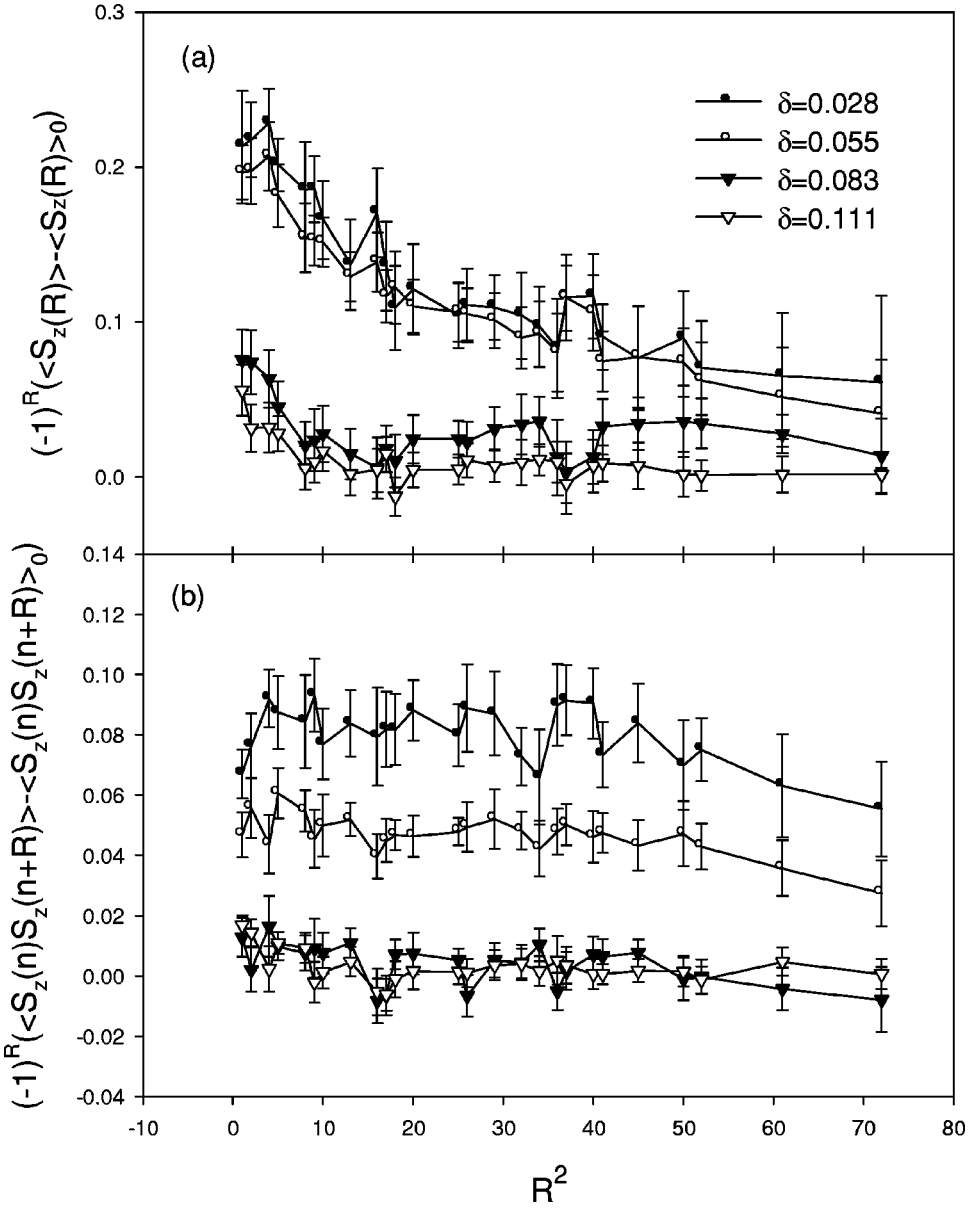

FIG. 2. Enhancement of (a) S_z and (b) spin-spin correlation function for diffferent hole densities plotted as a function of the square of the distance from the impurity.

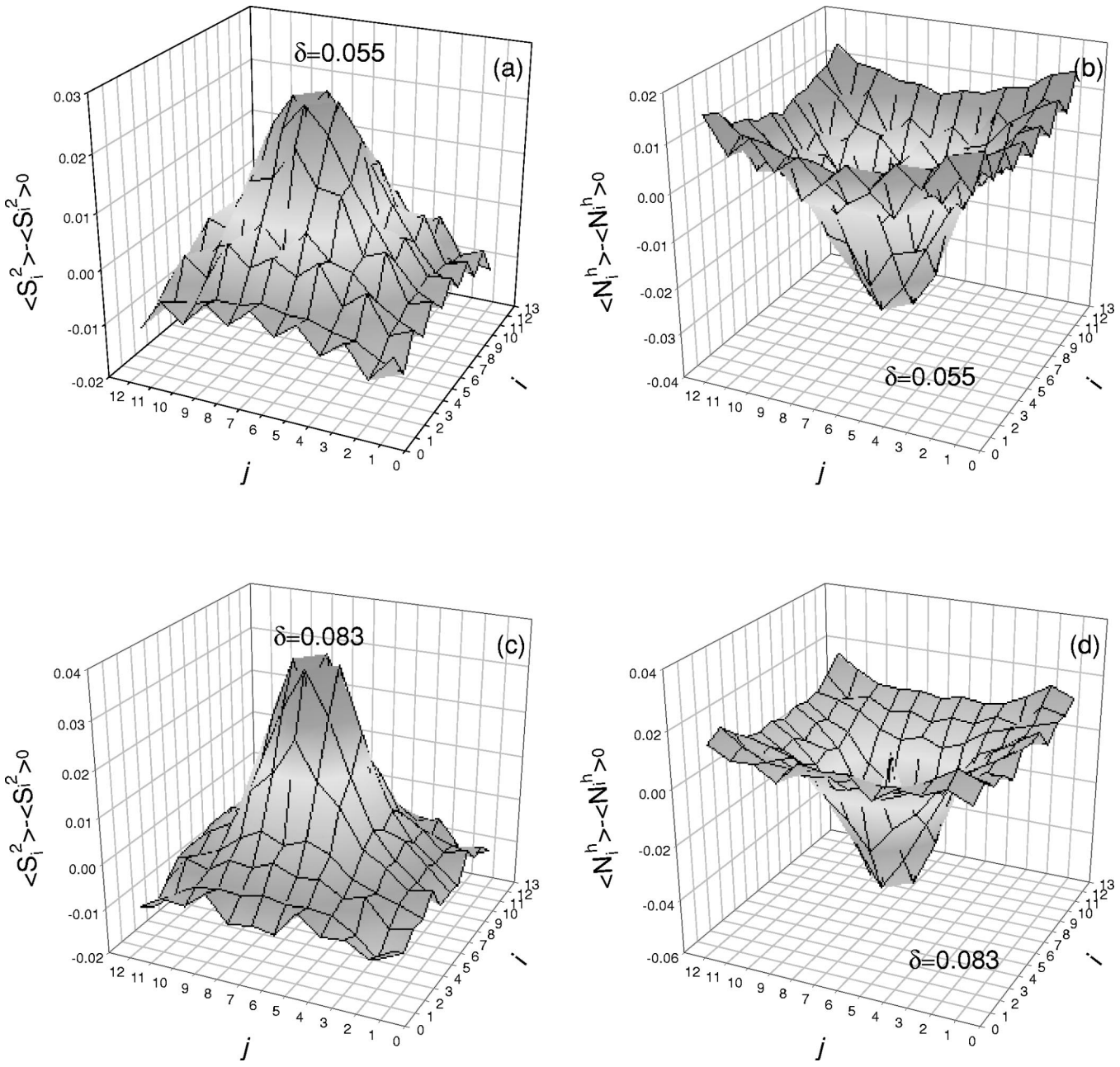

FIG. 3. Spin and charge profiles for different hole densities calculated by $|\psi_I\rangle$. The parameters are listed in Table II.

achieved for $h_u=0.0$, $h_0=0.0$, and $\lambda=0.2$. It is noted that the energy is quite sensitive to the value of λ. The lowest energy is acquired for positive λ, thus the hole is repelled away from the impurity while the moment is bound to the impurity.

We compare the optimal ground-state energy per site calculated from the trial wave functions $|\phi\rangle$ and $|\psi_I\rangle$ in Table I. In the third row we also list the total energy difference (ΔE) between these two wave functions. The variational parameters for the optimized wave function are listed in Table II.

As shown in Table I the Jastrow factor that simulates the magnetic polarization around the impurity in Eq. (9) reduces the energy of the projected BdG wave function $|\phi\rangle$ by a significant amount. Although the energy per site has been improved only by a very small amount, the total energy gain is greater than $0.7J$. This is a very large energy gain due to the influence of a single impurity. It also clearly demonstrates that BdG approach has significantly underestimated the magnetic correlation surrounding the impurity.

Table II shows that h_u is zero, i.e., there is no AF LRO for doping greater than 0.08 with or without the impurity. This is expected as a single impurity cannot induce LRO for the whole system. At the underdoped region, for $\delta=0.083 \sim 0.11$, although there is no uniform AF LRO, the spins around the impurity tend to form a local AF cloud as reflected by the nonvanishing parameter h_0. It should be cautioned that in this case our trial function $|\psi_I\rangle$ has broken the spin up-down symmetry. A more accurate description of this state should be a state with a fluctuating local AF polarization but without fixing the moment in a particular direction.

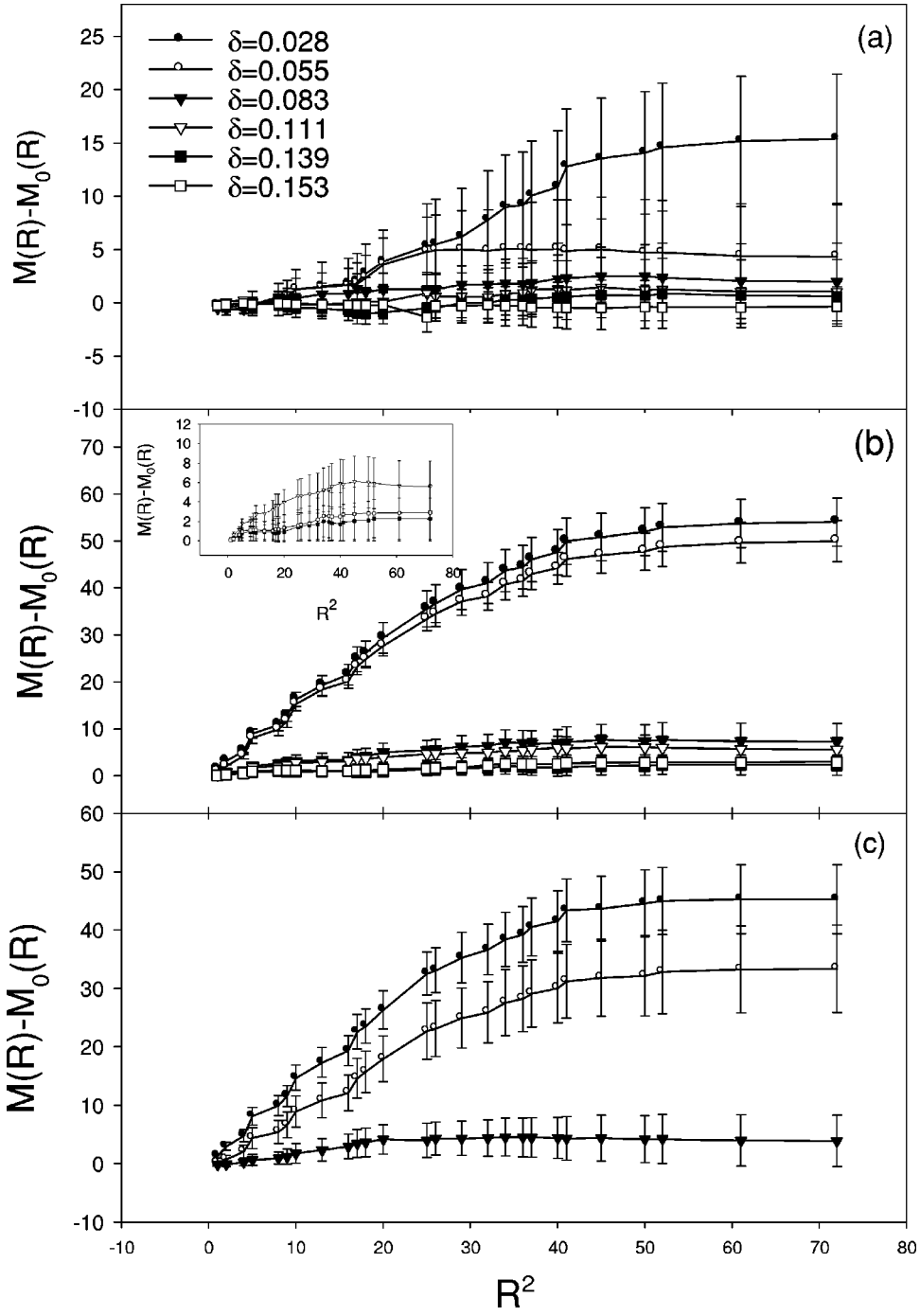

FIG. 4. The induced magnetic polarization $M(R)-M_0(R)$, plotted as a function of R^2 for different hole densities, obtained from (a) the BdG wave function $|\phi\rangle$ and (b) the wave function $|\psi_I\rangle$. The parameters are listed in Table II. In the inset, $\delta=0.111$, 0.139, and 0.152 are shown with a different scale; (c) is obtained from $|\psi_I'\rangle$ which is similar to $|\psi_I\rangle$ used in (b) but with a different functional form for the Jastrow factor as discussed in the text. The parameters used are $\delta=0.028$, $h_u=0.1$, $h_0=0.2$, $\lambda=1.2$; $\delta=0.055$, $h_u=0.05$, $h_0=0.1$, $\lambda=1.0$; and $\delta=0.083$, $h_u=0$, $h_0=0.025$, $\lambda=0.8$.

For $\delta\geqslant 0.139$ although $h_0=0$ and there is no apparent magnetic polarization around the impurity, the holes are still repelled from the impurity. This result disagrees with the result reported by Tsuchiura *et al.*[21]

To examine the magnetic polarization induced around the impurity more closely, we have calculated the difference of the local magnetization $\langle S_z(R)\rangle$ and the spin-spin correlation function $\langle S_z(n)S_z(n+R)\rangle$ between systems with and without impurity. Both results are plotted in Fig. 2 as a function of the square of the distance from the impurity for several dopant densities. $(-1)^R[\langle S_z(R)\rangle-\langle S_z(R)\rangle_0]$ shown in Fig. 2(a) indicates that $\langle S_z(R)\rangle$ is enhanced near the impurity. $\langle\cdots\rangle_0$ is for the clean system without impurity. For $\delta\geqslant 0.083$ there is no AF LRO and the induced magnetization only exists within a few lattice constants around the impurity. In Fig. 2(b) we show that the spin-spin correlation is also enhanced near the impurity. Site n is one of the nearest neighbors of the impurity. Again the enhancement is weaker when the doping increases. This is consistent with the experimental observation.

In Fig. 3 we plot the impurity-induced spin and charge profiles, $\Delta S^2=\langle S_i^2\rangle-\langle S_i^2\rangle_0$, and $\Delta N_h=\langle N_i^h\rangle-\langle N_i^h\rangle_0$, respectively, for two different dopant concentrations. Here $N_i^h=1-n_{i\sigma}-n_{i-\sigma}$. It can be seen that the holes are kept away from the impurity and a spin cloud is formed around the

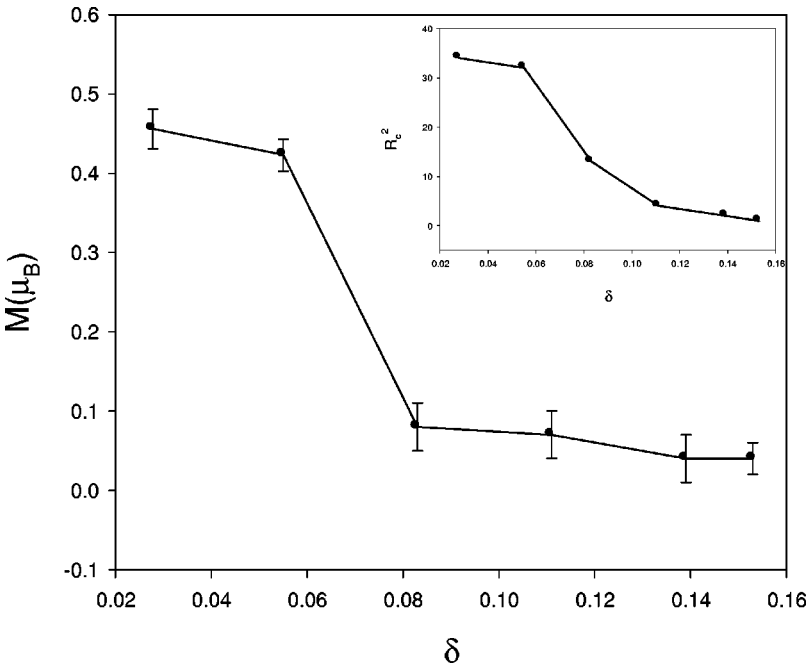

FIG. 5. The induced magnetic moment plotted as a function of hole density. In the inset, the size of the spin cloud versus hole density is shown. The parameters are listed in Table II.

impurity. As the hole doping increases the spin cloud becomes smaller in size.

To estimate the size of the induced magnetic polarization and the induced moment, we calculate $M(R) = 3g\langle\sqrt{(\Sigma_i^{N_R}(-1)^i S_i^z)^2}\rangle$, where the Lande g factor $g=2$ and N_R is the number of sites within radius R of the impurity. The difference between the induced magnetization with and without impurity, $M(R)-M_0(R)$, is plotted as a function of R^2 in Fig. 4. Results obtained by using $|\phi\rangle$ and $|\psi_I\rangle$ are shown in Figs. 4(a) and 4(b), respectively. In the inset of Fig. 4(b), results for $\delta=0.111$, 0.139, and 0.152 are shown with a different scale. The saturation of values of $M(R)-M_0(R)$ at large R indicates that the induced magnetization has a finite extent. We shall define the size of the induced magnetic polarization to be R_c. At $R=R_c$, $M(R)-M_0(R)$ reaches about 70% of its saturated values. This moment is much larger for $|\psi_I\rangle$ than for $|\phi\rangle$. Hence the local staggered magnetic field h_i and the repulsion between impurity and hole introduced by the Jastrow factor in Eq. (9) has enhanced the induced moment.

In Fig. 4(b) we have used $h_i=h_u+(h_0/R_i)$ and $1/R$ for the repulsion between hole and impurity in Eq. (9) for $|\psi_I\rangle$. To examine the sensitivity of the result to the choice of the R dependence, we change $1/R$ to $1/R^2$ for both h_i and the repulsion term in the Jastrow factor. The optimized variational energies are almost the same as the results reported in Table I. The results for the induced magnetic polarization are plotted in Fig. 4(c), which are quite similar to Fig. 4(b).

Results in Fig. 4 show that in the AF LRO states or $\delta \leqslant 0.083$, the induced magnetization is much larger. When there is no LRO the induced magnetization decreases rapidly with increasing hole concentration. This is consistent with experiments.[10] It is also consistent with the theoretical result reported by Tsuchiura *et al.*[21] But we do not agree with their conclusion that the holes are attracted toward the impurity. On the contrary, we have shown above that the holes are repelled away from the impurity to lower their kinetic energy. This effect might give an explanation to the similarity[3] between Li^+ and Zn^{2+}. The holes are also repelled away from the Li to gain energy.

The induced moment $M=[M(R_c)-M_0(R_c)]/N_{R_c}$ and the square of the size of the induced cloud, R_c^2, are plotted as a function of hole concentration in Fig. 5 and its inset, respectively. For the hole concentration $\delta \leqslant 0.055$, the local magnetic moment we obtained is about $0.5\mu_B$ as compared with the experimental value $0.4-1.0\mu_B$ for the Zn 4% substitution and different dopings. The rapid decrease of the size of the induced spin cloud could be due to the screening by the conducting carriers.[3,17]

In summary, the magnetic polarization induced by nonmagnetic impurities in high-T_c cuprate compounds is studied by combining the variational Monte Carlo simulation and Bogoliubov de Gennes mean-field Hamiltonian for the two-dimensional t-J model. A Jastrow factor is introduced to account for the induced magnetic moment and the repulsion between holes and the impurity. A substantial energy gain is obtained when the holes are repelled and the antiferromagnetic polarization is enhanced near the impurity. The doping dependence for the induced magnetic moment is consistent with experiments.

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